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Mar 5, 1999

DERWENT-ACC-NO: 1999-233140
DERWENT-WEEK: 199920
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TITLE: Copper-carbon fiber composite for heat dissipation substrate of semiconductor devices etc. - consists of carbon fibers in copper matrix with compound of carbon and copper having higher melting point than copper, at boundary surface of composite

PATENT-ASSIGNEE: HITACHI LTD (HITA)

PRIORITY-DATA: 1997JP-0232104 (August 28, 1997)

PATENT-FAMILY:

PUB-NO	PUB-DATE	LANGUAGE	PAGES	MAIN-IPC
JP <u>11061292</u> A	March 5, 1999		007	C22C001/09

APPLICATION-DATA:

PUB-NO	APPL-DATE	APPL-NO	DESCRIPTOR
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INT-CL (IPC): B22 F 3/14; C22 C 1/09; C22 C 9/00; H01 L 23/14

ABSTRACTED-PUB-NO: JP11061292A
BASIC-ABSTRACT:

NOVELTY - Carbon fibers are contained in a copper matrix comprising chiefly of copper. The boundary surface of the composite includes a compound of carbon and copper which has a melting point higher than that of copper.

DETAILED DESCRIPTION - An INDEPENDENT CLAIM is also included for the copper-carbon fiber composite manufacturing method.

USE - For heat dissipation substrate of semiconductor devices.

ADVANTAGE - Composite having low thermal expansion and high thermal conductivity is obtained.

ABSTRACTED-PUB-NO: JP11061292A
EQUIVALENT-ABSTRACTS:

CHOSEN-DRAWING: Dwg.1/8

DERWENT-CLASS: L03 M22 P53 U11
CPI-CODES: L04-C22; M22-G03K;
EPI-CODES: U11-D02B;

COPPER-CARBON FIBER COMPOSITE AND ITS PRODUCTION

Patent Number: JP11061292
Publication date: 1999-03-05
Inventor(s): KANEDA JUNYA; TAKASE IWAO; GOTO SUMITAKA; KAMOSHITA RIKUO;
INAGAKI MASATOSHI
Applicant(s):: HITACHI LTD
Requested Patent: ☐ JP11061292
Application
Number: JP19970232104 19970828
Priority Number
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IPC Classification: C22C1/09 ; B22F3/14 ; C22C9/00 ; H01L23/14
EC Classification:
Equivalents:

Abstract

PROBLEM TO BE SOLVED: To obtain a copper-carbon fiber composite having a low coefft. of expansion and high heat conductivity by incorporating a specified metallic element into the interfaces between a copper matrix and carbon fibers and allowing specified interfacial layers to exist.
SOLUTION: The copper-carbon fiber composite contains a metallic element having a higher m.p. than copper and forming a compd. with carbon in the interfaces between the copper matrix made substantially of only copper and carbon fibers and has interfacial layers formed by allowing the metallic element to react with Cu and/or C or to enter into solid soln. in Cu and/or C. The metallic element is preferably Si, Ti, V, Cr, Zr, Nb, Mo, Hf, Ta or W. The carbon fiber content of the composite is preferably 20-70 vol.%. In the case of <20 vol.%, thermal expansion suppressing effect is low. In the case of >70 vol.%, heat conductivity is reduced. The average thickness of the interfacial layers is preferably $\leq 2 \mu\text{m}$. In the case of $>2 \mu\text{m}$, heat conductivity is reduced. The composite is produced by mixing Cu powder with carbon fibers and powder of the metallic element and press-compacting the mixture under heating in a short time.

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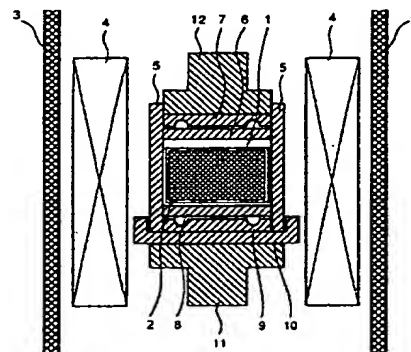
(54) 【発明の名称】 銅-炭素繊維複合体及びその製造方法

(57) 【要約】

【課題】 銅マトリックス中に炭素繊維を含有した低熱膨張、高熱伝導の銅-炭素繊維複合体及びその製造法を提供する。

【解決手段】 前記銅-炭素繊維複合体の銅マトリックスと炭素繊維の界面に炭素と化合物を形成する金属元素を含み、かつ銅あるいは炭素またはその両方に対して反応又は固溶した界面層が存在し、銅マトリックスは実質的に銅からなるものである。

図 1



【特許請求の範囲】

【請求項1】銅マトリックス中に炭素繊維を含有する銅-炭素繊維複合体において、銅マトリックスが不可避免の不純物元素を除き実質的に銅から成り、銅と炭素繊維との界面に金属元素を有し、該金属元素が炭素あるいは銅またはその両方に対して反応又は固溶した界面層が存在することを特徴とする銅-炭素繊維複合体。

【請求項2】銅マトリックス中に炭素繊維を含有する銅-炭素繊維複合体において、銅マトリックスが不可避免の不純物元素を除き実質的に銅から成り、銅と炭素繊維との界面に銅より融点が高く、炭素と化合物を形成する金属元素を有し、該金属元素が前記炭素あるいは銅またはその両方に対して反応又は固溶した界面層を有することを特徴とする銅-炭素繊維複合体。

【請求項3】銅マトリックス中に炭素繊維を含有する銅-炭素繊維複合体において、銅マトリックスが不可避免の不純物元素を除き実質的に銅から成り、銅と炭素繊維との界面にSi, Ti, V, Cr, Zr, Nb, Mo, Hf, Ta, Wのうちの少なくとも一つの金属元素を含み、該金属元素が前記炭素あるいは銅またはその両方に対して反応又は固溶した界面層を有することを特徴とする銅-炭素繊維複合体。

【請求項4】請求項1～3のいずれかにおいて、前記金属元素はその重量比でMm、その比重を ρ 、炭素の含有量を重量比でMcfとしたときに、 $Mm/(Mcf \times \rho)$ の値が0.1以下であることを特徴とする銅-炭素繊維複合体。

【請求項5】請求項1～4のいずれかにおいて、前記炭素繊維はその体積率で20～70%であることを特徴とする銅-炭素繊維複合体。

【請求項6】請求項1～4のいずれかにおいて、前記界面層の厚さが2.0 μ m以下であることを特徴とする銅-炭素繊維複合体。

【請求項7】請求項1～6のいずれかにおいて、熱膨張率が $12 \times 10^{-6}/^{\circ}\text{K}$ 以下及び熱伝導度が $120\text{W}/\text{m}^{\circ}\text{K}$ 以上であることを特徴とする銅-炭素繊維複合体。

【請求項8】銅マトリックス中に炭素繊維を含有する銅-炭素繊維複合体の製造方法において、銅粉末、炭素繊維及び該炭素繊維と化合物を形成する金属元素粉末を混合した後、加熱加圧成形し、前記銅マトリックスが不可避免の不純物元素を除き実質的に銅から成り、銅と炭素繊維との界面に炭素あるいは銅またはその両方に対して反応又は固溶した界面層を形成させることを特徴とする銅-炭素繊維複合体の製造方法。

【請求項9】銅マトリックス中に炭素繊維を含有する銅-炭素繊維複合体の製造方法において、銅粉末、炭素繊維及び銅より融点が高く、炭素と化合物を形成する金属元素粉末を混合した後、加熱加圧成形し、

前記銅マトリックスが不可避免の不純物元素を除き実質的に銅から成り、銅より融点が高く、炭素と化合物を形成する前記金属元素粉末が前記炭素あるいは銅またはその両方に対して反応又は固溶した界面層を形成させることを特徴とする銅-炭素繊維複合体の製造方法。

【請求項10】銅マトリックス中に炭素繊維を含有する銅-炭素繊維複合体の製造方法において、銅粉末、炭素繊維及びSi, Ti, V, Cr, Zr, Nb, Mo, Hf, Ta, W粉末のうちの少なくとも一つの金属元素粉末を混合した後、加熱加圧成形し、

前記銅マトリックスが不可避免の不純物元素を除き実質的に銅から成り、前記金属元素粉末が前記炭素あるいは銅またはその両方に対して反応又は固溶した界面層を形成させることを特徴とする銅-炭素繊維複合体の製造方法。

【請求項11】銅マトリックス中に炭素繊維を含有する銅-炭素繊維複合体において、該複合体の熱膨張率が $12 \times 10^{-6}/^{\circ}\text{K}$ 以下及び熱伝導率が $120\text{W}/\text{m}^{\circ}\text{K}$ 以上であることを特徴とする銅-炭素繊維複合体。

【請求項12】半導体素子を熱伝導性基板上に搭載した半導体装置において、前記基板は請求項1～7及び11のいずれかに記載の銅-炭素繊維複合体からなることを特徴とする半導体装置。

【発明の詳細な説明】

【0001】

【発明の属する技術分野】本発明は、新規な銅-炭素繊維複合材に係わり、特に半導体装置用放熱基板等として用いられる低熱膨張でかつ高熱伝導性の銅-炭素繊維複合材料及びその製造方法に関する。

【0002】

【従来の技術】銅は熱伝導度が大きく、広く放熱用の材料として用いられている。しかし、熱膨張率が $17 \times 10^{-6}/^{\circ}\text{K}$ と大きいために半導体装置用放熱基板等の低熱膨張性が要求される部材に用いることが出来ない。現在、高密度半導体装置用放熱基板にはモリブデンが用いられているが、より熱伝導度が高く、低コストの材料が望まれている。これまでに、低熱膨張率の炭素繊維と高熱伝導性の銅の複合化による、低熱膨張でかつ高熱伝導の複合材の開発がなされてきた。その製造方法には、

(1) 銅を溶湯にして炭素繊維に含浸させる方法、

(2) 板あるいは箔状の銅と炭素繊維を交互に積層してホットプレスする方法、(3) 炭素繊維に銅を蒸着あるいはメッキにより付着させた後ホットプレスする方法がある。(1)の方法は、銅と炭素繊維の濡れ性が悪いことから、炭素繊維の間隔の狭い複合体を得ようとするときには、炭素繊維間に銅が含まれず、空洞が形成される。(2)の方法においても、(1)と同様な現象が生じると考えられ、材料強度上好ましくない。(3)の方法では、高度な蒸着あるいはメッキ技術が要求されるという問題点が挙げられる。

【0003】このような問題を解決するために、様々な方法により銅-炭素繊維複合体の製造方法が開発されている。特開昭51-5213号は、銅あるいは銅合金の粉末を粘結剤と混合してスラリーとし、炭素繊維束に含浸させた後、プレスや焼結により所定の形状に固化する方法である。特開昭52-89505号、特開昭52-53720号は、銅あるいは炭素と反応する元素が固溶限以上添加された銅合金を、軟化点以上の温度で加圧することにより炭素繊維に含浸させ複合体を形成する方法である。また、特開昭53-104508号は粉末状高導電性金属、炭素粉、炭素と反応する元素粉末、粘結剤を混合してスラリーとし、これを高導電性金属被覆炭素繊維に含浸させた後、熱間加圧成形あるいは溶解冷却成形を行い複合体を製造する方法である。

【0004】

【発明が解決しようとする課題】これらの製造方法で得られた銅-炭素繊維複合体は、熱膨張率がモリブデンと同等に小さいものは得られているが、熱伝導率が低く、モリブデンに代替するには至っていない。この原因は、炭素繊維密度の増加と炭素と反応する元素の添加にある。本来、銅と炭素とは互いに非固溶であり、濡れ性が悪く、複合体と成り難い材料である。高熱伝導度、低熱膨張率、高強度の銅-炭素繊維複合体は、銅マトリックスと炭素繊維間の結合により炭素繊維が銅マトリックスの熱膨張を抑え、かつ銅マトリックス中の熱伝導により高い熱伝導率が維持されることが必要である。また、複合体の内部に空洞や割れ等の材料強度上好ましくない要因がないことも必要である。しかし、一般に銅と炭素繊維の界面の結合力が弱いために炭素繊維が銅の熱膨張を抑制できず、また濡れ性が悪いために銅の含浸が不十分となり空洞が形成され易い。この問題を解決するために、炭素と反応する元素を添加することにより銅と炭素繊維の濡れ性を改善し結合力を向上させた複合体が作製されている。しかし、熱膨張率は小さくなるが、熱伝導を担う銅マトリックス中に添加元素が固溶するために十分な熱伝導度を得ることが出来ない。また、熱膨張率は単位体積当たりの炭素繊維本数が多くなるに従い低下するが、これによっても熱伝導を担う銅の体積率が減少するために熱伝導度が低下する。従って、銅-炭素繊維複合体においては、熱膨張率の抑制と熱伝導度の向上を同時に達成することが困難であった。

【0005】本発明は、低熱膨張率と高熱伝導率を同時に満たす銅-炭素繊維複合体及びその製造方法を提供することを目的とする。

【0006】

【課題を解決するための手段】本発明は、銅マトリックスが不可避的不純物元素を除き実質的に銅のみから成り、銅と炭素繊維の界面に銅あるいは炭素またはその両方に対して反応又は固溶した界面層が存在する組織を有する銅と炭素繊維複合体にある。即ち、銅と炭素繊維の

結合力を向上させた銅-炭素繊維複合体が有効であることを見出した。前記界面層は、銅より融点が高かつ炭素と化合物を形成する金属元素を含まなければならない。このような炭素化合物形成金属元素としては、Si, Ti, V, Cr, Zr, Nb, Mo, Hf, Ta, Wが好ましく、これらのうちの少なくとも一つの元素が炭素繊維と銅マトリックスの界面に界面層を形成している必要がある。また、銅と炭素繊維の界面にSi, Ti, Zr, Hfのうちの少なくとも一つの元素を含む界面層が存在することがより好ましい。上記の金属元素添加により、炭素繊維と銅マトリックス間の結合状態が改善されても、銅マトリックス中に上記金属元素が固溶すると、複合体の熱伝導度を低下させる。そこで、銅マトリックスは不可避的不純物元素を除き実質的に銅のみから成るように製造するものでなければならない。

【0007】上記の銅-炭素繊維複合体において、炭素繊維体積分率が小さいと熱膨張の抑制効果が小さく、また炭素繊維体積分率が大きいと熱伝導度が小さくなる。このため、炭素繊維体積分率は20%以上70%以下であることが好ましい。また、界面層の厚さが厚い場合には、隣接する炭素繊維間に界面層のみが存在し、銅が含浸されないために、熱伝導度が低下する。このため、界面層の平均厚さは2 μ m以下が好ましい。界面層の厚さを調整し、かつ銅マトリックス中に固溶する添加金属元素濃度を抑えるためには、炭素繊維重量をMcf、炭素と化合物を形成する金属元素の重量をMm、炭素と化合物を形成する金属元素の比重を ρ としたときに、銅-炭素繊維複合体中に含まれる、炭素と化合物を形成する金属元素それぞれについての $Mm/(Mcf \times \rho)$ の値の和が0.1以下であることが必要である。このような材料組織を有する銅-炭素繊維複合体は、熱膨張率 $12 \times 10^{-6}/^{\circ}\text{K}$ 以下、熱伝導度 $120\text{W}/\text{m}^{\circ}\text{K}$ 以上を達成出来る。

【0008】上記の銅-炭素繊維複合体は、銅粉末、炭素繊維及び炭素と化合物を形成することが可能な金属元素粉末を混合し、短時間で加熱加圧成形し、銅マトリックスが実質的に銅からなり、前述の界面層を形成させることを特徴とする銅-炭素繊維複合体の製造方法により製造することが出来る。

【0009】上記の銅-炭素繊維複合体を、半導体素子を搭載した半導体装置の熱伝導性基板として用いることにより、半導体装置の品質において信頼性が高く、放熱性に優れた半導体装置を提供することが出来る。

【0010】本発明は、低熱膨張で、かつ高熱伝導性の銅-炭素繊維複合体を提供する。銅と炭素繊維とは、互いに非固溶であり、濡れ性が悪く、複合体を作製し難い。また、複合体を作製しても熱膨張を充分に抑制出来なかつたり、また熱伝導度を低下させたりする。そこで、銅マトリックスと炭素繊維の間に炭素と化合物を形成することの出来る金属元素を含む界面層を形成するこ

とにより、銅マトリックスと炭素繊維の結合力を向上させ、所定の熱膨張率及び熱伝導度を達成することが出来た。ここでは、炭素と化合物を形成することの出来る金属元素としては、周期率表で4A、5A、6A族の元素及びSi、すなわちSi、Ti、V、Cr、Zr、Nb、Mo、Hf、Ta、Wが好ましい。特に、界面層形成及び銅マトリックスとの反応の点でSi、Ti、Zr、Hfが好ましい。また、優れた熱伝導性を維持する上で、銅マトリックスは、不可避的不純物元素を除き実質的に銅のみから成っていないなければならない。特に、銅-炭素繊維複合体に添加されている、炭素と化合物を形成することの出来る金属元素は、銅マトリックス中に含まれていないことが好ましいが、銅マトリックス中に含まれる上記添加元素の定量分析分解能に限界があることから、分析により検出される上記添加元素濃度は0.5wt.%以下とする。

【0011】本発明の銅-炭素繊維複合体の熱膨張率は、主に炭素繊維の体積分率に依存し、また、熱伝導度は主に炭素繊維体積分率及び銅マトリックスへの添加元素の固溶量に依存する。放熱用基板に用いるためには、熱膨張率 $12 \times 10^{-6} / ^\circ\text{K}$ 以下及び熱伝導度 $120\text{W} / \text{m}^\circ\text{K}$ 以上が必要である。

【0012】炭素繊維体積分率は、銅-炭素繊維複合体の熱膨張率、熱伝導度共に影響を及ぼす。炭素繊維体積分率と熱膨張率及び熱伝導度の関係から、上記特性を達成するのは、炭素繊維体積分率が20～70%のときであった。しかし、炭素繊維の種類や銅-炭素繊維複合体製造時の条件のばらつき等を考慮すると、炭素繊維体積分率が30～60%が好ましい。

【0013】銅マトリックスと炭素繊維界面に形成されている界面層は、その厚さが増すと隣接する炭素繊維間には浸透されて熱伝導を担うべき銅の量が減少し、所定の熱伝導度を得ることが出来なくなる可能性がある。そこで、炭素繊維体積分率を約50%に固定して、炭素化合物を形成するための添加金属元素量を変化させて、界面層厚さの異なる銅-炭素繊維複合体を作製し、その熱伝導特性を評価した。その結果、界面層厚さが平均 $2\mu\text{m}$ 以下であれば、目標の熱伝導度を保持できることが確認された。より薄い界面層となれば、より高い熱伝導度を達成することが出来ることから、原子層レベルでの炭素化合物の形成が理想的であるが、銅-炭素繊維複合体製造の点で、 $0.05\mu\text{m}$ 程度が限界であると考えられることから、平均 $0.05 \sim 1\mu\text{m}$ の界面層厚さが好ましい。

【0014】炭素化合物形成のための添加金属元素量は、上記のように界面層厚さに影響を与えるだけでなく、銅マトリックス中に固溶して、銅マトリックスの熱伝導度を低下させる。そこで、添加金属元素量と熱伝導度の関係を調査した。その結果、目標の熱伝導度を達成するためには、銅-炭素繊維複合体中の添加金属元素重

量比をMm、炭素繊維重量比をMcf、添加金属元素の比重を ρ としたとき $Mm / (Mcf \times \rho)$ の値が0.1以下であることが必要である。特に、銅マトリックス中への固溶量を低減させるため、及び実質的に均一な界面層を得るためには、 $Mm / (Mcf \times \rho)$ の値は0.005以上で0.08以下が好ましい。

【0015】上記のような材料組織及び特性を有する銅-炭素繊維複合体は、銅粒子、炭素繊維及び炭素と化合物を形成することが可能な金属元素粒子を混合し、加熱、加圧成形して作製した。上記の炭素と化合物を形成することが可能な金属元素は、具体的にはSi、Ti、V、Cr、Zr、Nb、Mo、Hf、Ta、Wである。これらのうち、特にSi、Ti、Zr、Hfが好ましい。これらの金属元素は0.2～5重量%、より0.5～3重量%が好ましい。また、上記の加熱、加圧成形は、真空中が好ましいが、銅マトリックス中への酸素の固溶を抑制出来る減圧あるいは不活性ガス雰囲気でもよい。加熱温度は、銅の融点以上であることが好ましいが、 800°C 以上であればよい。加圧力は加熱温度に依存するので、加熱温度に応じた加圧力を加える必要がある。この方法によれば、上記金属元素の銅中への固溶を抑え、かつ銅と炭素繊維界面に良好な界面層を形成することが出来る。

【0016】このような製造方法で作製された、所定の組織及び特性を有する銅-炭素繊維複合体は、低熱膨張及び高熱伝導が必要とされる部位に適用することが出来る。特に、銅-炭素繊維複合体と窒化アルミ及びアルミナの板との銀ろうによる接合が可能であることから、半導体装置の熱伝導性基板に適用することが出来る。

【0017】

【発明の実施の形態】

【実施例1】銅-炭素繊維複合体の製造方法を示す。

【0018】ピッチ系炭素繊維を2～6mmの長さに切断し、これに直径 $10\mu\text{m}$ 以下のTi粒子を混合した。更に、その炭素繊維とTi粒子混合物とに100メッシュ以下のCu粒子を混合した。混合重量比は、炭素繊維：Cu粒子：Ti粒子=9.54(g)：36.4(g)：2.05(g)であった。また、一連の混合により炭素繊維は切断されたままの状態より多少粉碎された。炭素繊維、Cu粒子、Ti粒子の混合物1を 0.1mm^2 のCu板で作った箱(Cu容器)2の中に入れ、図1に示す加熱加圧成形装置に入れた。ただし、図1では、加熱加圧成形装置の断面を示している。加熱加圧成形装置には雰囲気制御のためにチャンバー3が設けられており、その中には加熱炉4が設置されている。混合物1は、側面黒鉛治具5、上部黒鉛板6、上部湯だめ付き黒鉛板7、下部黒鉛板8、下部湯だめ付き黒鉛板9で包囲した後、湯流れ防止皿10にのせて、加熱炉4内に設置した。このとき、加圧のための下部加圧治具11、上部加圧治具12は、混合物1が加熱炉4の中央に位置するように調節

した。上記の状態に設置、調節した後、チャンバー3内を 1×10^{-5} Torr以上の真空度とした。なお、Cu容器2と黒鉛治具の隙間にはCu粒を充填した。

【0019】この後、混合物1に加圧治具にて 20 kg/cm^2 の圧力をかけた状態で、 1100°C 、 10 min の加熱加圧成形を実施した。これにより、直径 6.5 mm 、厚さ 4 mm の円板上の銅-炭素繊維複合体を作製した。

【0020】このようにして作製した銅-炭素繊維複合体の走査型電子顕微鏡(SEM)によって観察した結果、黒いコントラストの部分が炭素繊維、白いコントラストの部分が銅マトリックス、そして、炭素繊維と銅マトリックスの界面に存在する灰色の界面層が観察された。SEMに備えられている元素分析装置(EDX)で炭素繊維、界面層、銅マトリックスの各部分を分析し、図2、図3、図4に示す結果を得た。これより、炭素繊維と銅マトリックスの界面にTi、Cu、炭素を含む界面層が形成されていることがわかる。また、銅マトリックス中には、実質的に銅からのX線ピークしか観察されていない。作製した銅-炭素繊維複合体の各元素重量比は、 $\text{Cu}:\text{Ti}:\text{C}=87.2:2.6:10.2$ であった。

【0021】(実施例2) 実施例1に示す銅-炭素繊維複合体と同じ製造方法により、約30、50、70%の炭素繊維体積分率となるように銅-炭素繊維複合体を作製した。このとき炭素繊維とTi粒子の重量比はほぼ4:1となるようにした。作製した円板状銅-炭素繊維複合体を室温 $\sim 300^\circ\text{C}$ の間で熱膨張率を測定した。また、熱伝導度をレーザーフラッシュ法で測定した。得られた熱膨張率及び熱伝導度と炭素繊維体積分率の関係をそれぞれ図5、図6に示す。これらより、 12×10^{-6}

表 1

添加金属元素	熱膨張率 ($\times 10^{-6}/^\circ\text{C}$)	熱伝導度 ($\text{W/m}^\circ\text{C}$)
Si	7.2	175
Zr	8.4	212
Hf	8.7	214

【0026】(実施例5) 実施例1で作製した直径 6.5 mm 、厚さ 4 mm の銅-炭素繊維複合体上に、銀ろうを用いて、一辺 40 mm 、厚さ 1 mm の正方形の窒化アルミ板を接合した。接合は、 800°C で 3 min 保持し、 $3^\circ\text{C}/\text{min}$ の冷却速度で冷却した。この接合方法では、銅-炭素繊維複合体と窒化アルミの接合体に割れは観察されなかった。また、反りもなかったことから、良好な接合状態であると言える。上記銅-炭素繊維複合体、炭化アルミ接合体を $-50 \sim 125^\circ\text{C}$ での熱サイクル試験を実施した。その結果、300サイクルまでは、接合体の劣化は観察されなかった。

【0027】

【発明の効果】本発明によれば、低コストで低熱膨張、

$^\circ\text{C}$ 以下の熱膨張率と $120 \text{ W/m}^\circ\text{C}$ 以上の熱伝導度を共に達成するには、炭素繊維体積分率は20~70%が適当である。

【0022】(実施例3) 炭素繊維体積分率を約50%に固定して、炭素繊維とTi粒子の重量比がほぼ4:1、2:1、1:1となるように配合して、実施例1と同じ製造方法を用いて銅-炭素繊維複合体を作製した。作製した銅-炭素繊維複合体の熱伝導度をレーザーフラッシュ法で測定した。また、Tiを含む界面層の平均厚さは、SEM写真より測定した。そこで、図7に示す界面層厚さと熱伝導度の関係を得た。これより、 $120 \text{ W/m}^\circ\text{C}$ 以上の熱伝導度を得るためには、 $2 \mu\text{m}$ 以下の界面層厚さが適当である。

【0023】また、銅-炭素繊維複合体に混合したTiと炭素繊維の重量比と熱伝導度の関係を整理すると、Ti重量をMm、炭素繊維重量をMcf、金属Tiの比重を ρ としたとき、図8の関係を得た。これより、 $120 \text{ W/m}^\circ\text{C}$ 以上の熱伝導度を得るためには、 $\text{Mm}/(\text{Mcf} \times \rho)$ の値が0.1以下が適当である。

【0024】(実施例4) 炭素繊維体積分率を約50%、 $\text{Mm}/(\text{Mcf} \times \rho)$ の値がほぼ0.05と成るようにSi、Zr、Hfをそれぞれ配合して、実施例1と同じ方法で銅-炭素繊維複合体を作製した。これらの熱膨張率、及び熱伝導度は表1に示す値となった。これより、Si、Zr、Hfは、Tiと同様に炭素繊維と銅マトリックスの界面に界面層を形成する効果が期待できる。

【0025】

【表1】

高熱伝導の銅-炭素繊維複合体が得られる。また、本発明の銅-炭素繊維複合体を半導体装置の熱伝導性基板として用いることにより、半導体装置の品質において信頼性が高く、放熱性に優れた半導体装置が得られる。

【図面の簡単な説明】

【図1】加熱加圧成形装置の断面図。

【図2】銅-炭素繊維複合体の炭素繊維部分のEDX分析結果。

【図3】銅-炭素繊維複合体の界面層部分のEDX分析結果。

【図4】銅-炭素繊維複合体の銅マトリックス部分のEDX分析結果。

【図5】銅-炭素繊維複合体の炭素繊維体積分率と熱膨

張率との関係を示す線図。

【図6】銅-炭素繊維複合体の炭素繊維体積分率と熱伝導度の関係を示す線図。

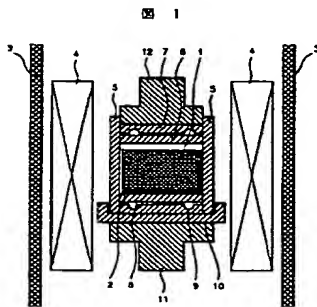
【図7】銅-炭素繊維複合体の炭素化合物層厚さと熱伝導度の関係を示す線図。

【図8】銅-炭素繊維複合体の金属添加量と熱伝導度の関係を示す線図。

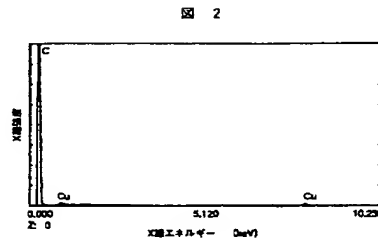
【符号の説明】

1…混合物、2…Cu容器、3…チャンパー、4…加熱炉、5…側面黒鉛治具、6…上部黒鉛板、7…上部湯だめ付き黒鉛板、8…下部黒鉛板、9…下部湯だめ付き黒鉛板、10…湯流れ防止皿、11…下部加圧治具、12…上部加圧治具。

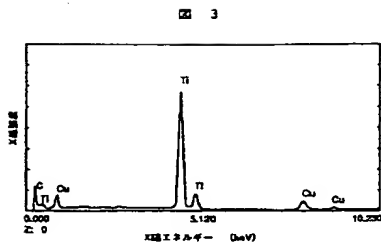
【図1】



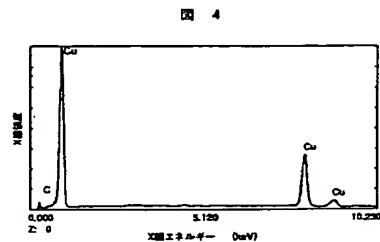
【図2】



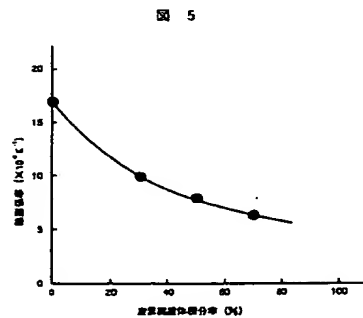
【図3】



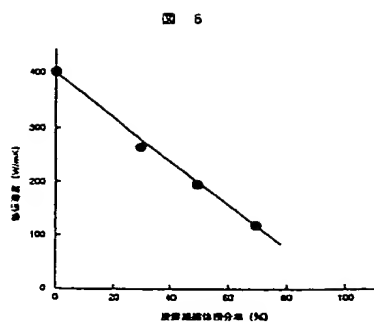
【図4】



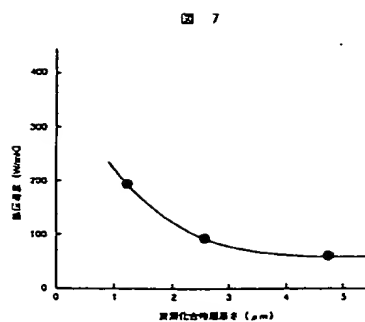
【図5】



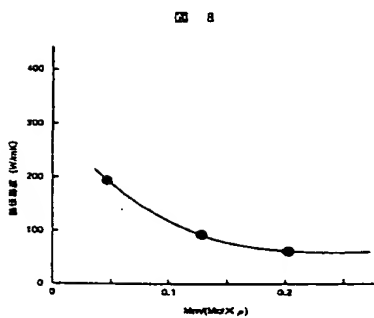
【図6】



【図7】



【図8】



フロントページの続き

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(54) COPPER-CARBON FIBER COMPOSITE AND ITS PRODUCTION

(57)Abstract:

PROBLEM TO BE SOLVED: To obtain a copper-carbon fiber composite having a low coefft. of expansion and high heat conductivity by incorporating a specified metallic element into the interfaces between a copper matrix and carbon fibers and allowing specified interfacial layers to exist.

SOLUTION: The copper-carbon fiber composite contains a metallic element having a higher m.p. than copper and forming a compd. with carbon in the interfaces between the copper matrix made substantially of only copper and carbon fibers and has interfacial layers formed by allowing the metallic element to react with Cu and/or C or to enter into solid soln. in Cu and/or C. The metallic element is preferably Si, Ti, V, Cr, Zr, Nb, Mo, Hf, Ta or W. The carbon fiber content of the composite is preferably 20-70 vol.%. In the case of <20 vol.%, thermal expansion suppressing effect is low. In the case of >70 vol.%, heat conductivity is reduced. The average thickness of the interfacial layers is preferably $\leq 2 \mu\text{m}$. In the case of $>2 \mu\text{m}$, heat conductivity is reduced. The composite is produced by mixing Cu powder with carbon fibers and powder of the metallic element and press-compacting the mixture under heating in a short time.

LEGAL STATUS

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3. In the drawings, any words are not translated.

DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] With respect to a new copper-carbon fiber composite, this invention is the low-thermal expansion especially used as a thermolysis substrate for semiconductor devices etc., and relates to the copper-carbon fiber composite material and its manufacture method of high temperature conductivity.

[0002]

[Description of the Prior Art] Thermal conductivity of copper is large and it is used as a large material for thermolysis. However, since coefficient of thermal expansion is as large as $17 \times 10^{-6}/\text{degree K}$, it cannot use for the member as which low-thermal expansibility, such as a thermolysis substrate for semiconductor devices, is required. Although molybdenum is used for the thermolysis substrate for high-density semiconductor devices now, thermal conductivity is more high and material of a low cost is desired. So far, it is low-thermal expansion by composite-izing of the carbon fiber of a low-thermal expansion coefficient, and the copper of high temperature conductivity, and development of the composite of high temperature conduction has been made. There are the method of carrying out the laminating of the copper and the carbon fiber of the shape of the method of making (1) copper into a molten metal and infiltrating a carbon fiber, (2) boards, or a foil, by turns, and carrying out a hotpress, and the method of carrying out a back hotpress of having made copper adhering to (3) carbon fibers by vacuum evaporation or plating as the manufacture method. Since the method of (1) has the bad wettability of copper and a carbon fiber, when it is going to obtain complex with the narrow interval of a carbon fiber, copper cannot be sunk in between carbon fibers but a cavity is formed. In the method of (2), it is thought that the same phenomenon as (1) arises, and it is not desirable on material strength. By the method of (3), the trouble that advanced vacuum evaporation or plating technology is required is mentioned.

[0003] In order to solve such a problem, the manufacture method of copper-carbon fiber complex is developed by various methods. After JP,51-5213,A mixes copper or the powder of a copper alloy with a binder, considers as a slurry and infiltrates a carbon fiber bunch, it is the method of solidifying in a predetermined configuration by the press or sintering. JP,52-89505,A and JP,52-53720,A are the methods of infiltrating a carbon fiber and forming complex, when copper or carbon, and the element that reacts pressurize the copper alloy added more than **** at the temperature more than softening temperature. Moreover, after JP,53-104508,A mixes a powdered high conductivity metal, a carbon powder, carbon and the element powder which reacts, and a binder, considers as a slurry and infiltrates this into a high conductivity metallic-coating carbon fiber, it is the method of performing pressing between heat, or dissolution cooling fabrication, and manufacturing complex.

[0004]

[Problem(s) to be Solved by the Invention] On a par [coefficient of thermal expansion] with molybdenum, its thermal conductivity is low and the copper-carbon fiber complex obtained by these manufacture methods has come to substitute molybdenum for it, although the small thing is obtained.

This cause is in addition of an increase and carbon of carbon fiber density, and the element which reacts. Originally, copper and carbon do not dissolve mutually, and wettability is bad and they are complex and the material which cannot change easily. The copper-carbon fiber complex of high temperature conductivity, a low-fee expansion coefficient, and high intensity needs for a carbon fiber to suppress the thermal expansion of a copper matrix by combination between a copper matrix and a carbon fiber, and to maintain high thermal conductivity by heat conduction in a copper matrix. Moreover, it is also required for the interior of complex for there to be no factor which is not desirable on material strength, such as a cavity and a crack. However, generally, since the bonding strength of the interface of copper and a carbon fiber is weak, a carbon fiber cannot suppress a copper thermal expansion, and since wettability is bad, it becomes inadequate copper sinking in, and a cavity is easy to be formed. In order to solve this problem, the complex which has improved the wettability of copper and a carbon fiber and raised bonding strength is produced by adding carbon and the element which reacts. However, although coefficient of thermal expansion becomes small, since an alloying element dissolves in the copper matrix which bears heat conduction, sufficient thermal conductivity cannot be obtained. Moreover, although it falls as the carbon fiber number of coefficient of thermal expansion per unit volume increases, in order that the rate of volume of the copper which bears heat conduction also by this may decrease, thermal conductivity falls. Therefore, in copper-carbon fiber complex, it was difficult to attain simultaneously suppression of coefficient of thermal expansion, and improvement in thermal conductivity.

[0005] this invention aims at offering the copper-carbon fiber complex which fills simultaneously a low-fee expansion coefficient and high temperature conductivity, and its manufacture method.

[0006]

[Means for Solving the Problem] A copper matrix consists only of copper substantially except for an unescapable impurity element, and this invention is in copper, the copper which has the organization where the volume phase which reacted or dissolved to copper, carbon, or its both to the interface of a carbon fiber exists, and carbon fiber complex. That is, it found out that the copper-carbon fiber complex which raised the bonding strength of copper and a carbon fiber was effective. The aforementioned volume phase must contain the metallic element in which the melting point forms carbon and a compound highly from copper. As such a carbon compound formation metallic element, Si, Ti, V, Cr, Zr, Nb, Mo, Hf, Ta, and W are desirable, and at least one of elements of these needs to form the volume phase in the interface of a carbon fiber and a copper matrix. Moreover, it is more desirable that the volume phase which contains at least one element in Si, Ti, Zr, and Hf in the interface of copper and a carbon fiber exists. If the above-mentioned metallic element dissolves in a copper matrix by it even if the integrated state between a carbon fiber and a copper matrix is improved by the above-mentioned metallic element addition, the thermal conductivity of complex will be reduced. Then, you have to manufacture a copper matrix so that it may consist only of copper substantially except for an unescapable impurity element.

[0007] In the above-mentioned copper-carbon fiber complex, if the depressor effect of thermal expansion is small when a carbon fiber volume fraction is small, and a carbon fiber volume fraction is large, thermal conductivity will become small. For this reason, as for a carbon fiber volume fraction, it is desirable that it is [20% or more] 70% or less. Moreover, since only a volume phase exists and copper does not sink in between the adjoining carbon fibers when the thickness of a volume phase is thick, thermal conductivity falls. For this reason, the average thickness of a volume phase has desirable 2 micrometers or less. In order to stop the addition metallic element concentration which adjusts the thickness of a volume phase and dissolves in a copper matrix, when the specific gravity of the metallic element which forms Mm, carbon, and a compound for the weight of the metallic element which forms Mcf, carbon, and a compound for a carbon fiber weight is set to rho, it is required for the sum of the value of $Mm/(Mcf \times \rho)$ about each carbon and metallic element which forms a compound contained in copper-carbon fiber complex to be 0.1 or less. The copper-carbon fiber complex which has such a material organization can attain less than [coefficient-of-thermal-expansion $12 \times 10^{-6}/\text{degree K}$] and more than thermal conductivity 120 W/m degree K.

[0008] The metallic element powder which can form a copper powder, a carbon fiber and carbon, and a compound is mixed, heating pressing is carried out for a short time, a copper matrix consists of copper substantially, and the above-mentioned copper-carbon fiber complex can be manufactured by the manufacture method of the copper-carbon fiber complex characterized by making the above-mentioned volume phase form.

[0009] By using the above-mentioned copper-carbon fiber complex as a thermally conductive substrate of a semiconductor device in which the semiconductor device was carried, in the quality of a semiconductor device, it is reliable, and the semiconductor device excellent in thermolysis nature can be offered.

[0010] this invention is low-fever expansion, and offers the copper-carbon fiber complex of high temperature conductivity. It does not dissolve mutually, and wettability is bad and, as for copper and a carbon fiber, cannot produce complex easily. Moreover, even if it produces complex, thermal expansion cannot fully be suppressed, and thermal conductivity is reduced. Then, by forming the volume phase containing carbon and the metallic element which can form a compound between a copper matrix and a carbon fiber, the bonding strength of a copper matrix and a carbon fiber was able to be raised, and a predetermined coefficient of thermal expansion and predetermined thermal conductivity were able to be attained. Here, as carbon and a metallic element which can form a compound, the element of 4A, 5A, and 6A group and Si, i.e., Si, Ti, V, Cr, Zr, Nb, Mo, Hf, Ta, and W, are desirable at a periodic table. Especially, Si, Ti, Zr, and Hf are desirable in respect of a reaction with volume phase formation and a copper matrix. Moreover, when maintaining the outstanding thermal conductivity, the copper matrix must consist only of copper substantially except for the unescapable impurity element. the quantitative analysis of the above-mentioned alloying element contained in a copper matrix although not being contained in a copper matrix is desirable as for especially the carbon and the metallic element which can form a compound added by copper-carbon fiber complex -- since a limitation is in resolution, the above-mentioned alloying-element concentration detected by analysis is made below into 0.5wt(s).%

[0011] The coefficient of thermal expansion of the copper-carbon fiber complex of this invention mainly depends for thermal conductivity on the amount of dissolution of the alloying element to a carbon fiber volume fraction and a copper matrix depending on the volume fraction of a carbon fiber. In order to use for the substrate for thermolysis, less than [coefficient-of-thermal-expansion 12×10^{-6} /degree K] and more than thermal conductivity 120 W/m degree K are required.

[0012] As for a carbon fiber volume fraction, the coefficient of thermal expansion of copper-carbon fiber complex and thermal conductivity do influence. From the relation between a carbon fiber volume fraction, coefficient of thermal expansion, and thermal conductivity, the time of a carbon fiber volume fraction being 20 - 70% attained the above-mentioned property. However, when the kind of carbon fiber, dispersion of the conditions at the time of copper-carbon fiber complex manufacture, etc. are taken into consideration, 30 - 60% has a desirable carbon fiber volume fraction.

[0013] It may become impossible for the amount of the copper which it sinks in between the carbon fibers which adjoin if the thickness increases, and should bear heat conduction to decrease, and for the volume phase currently formed in the copper matrix and the carbon fiber interface to obtain predetermined thermal conductivity. Then, the carbon fiber volume fraction was fixed to about 50%, the amount of addition metallic elements for forming a carbon compound was changed, the copper-carbon fiber complex with which volume phase thickness differs was produced, and the heat-conduction property was evaluated. Consequently, when volume phase thickness was an average of 2 micrometers or less, it was checked that target thermal conductivity can be held. Although formation of the carbon compound in atomic-layer level is ideal since higher thermal conductivity can be attained if it becomes a thinner volume phase, it is 0.05 micrometers at the point of copper-carbon fiber complex manufacture. An average of 0.05-1 micrometer since it is thought that a grade is a limitation Volume phase thickness is desirable.

[0014] The amount of addition metallic elements for carbon compound formation dissolves in a copper matrix, and it not only affects volume phase thickness as mentioned above, but it reduces the thermal conductivity of a copper matrix. Then, the relation between the amount of addition metallic elements

and thermal conductivity was investigated. Consequently, in order to attain target thermal conductivity, when the specific gravity of Mcf and an addition metallic element is set [the addition metallic element weight ratio in copper-carbon fiber complex] to ρ for Mm and a carbon fiber weight ratio, the value of $Mm/(Mcf \times \rho)$ is 0.1. It is required to be the following. In order to reduce the amount of dissolution to the inside of a copper matrix especially, and in order to obtain a uniform volume phase substantially, as for the value of $Mm/(Mcf \times \rho)$, 0.08 or less are desirable at 0.005 or more.

[0015] a metallic element particle with possible the copper-carbon fiber complex which has the above material organizations and properties forming a copper particle, a carbon fiber and carbon, and a compound -- mixing -- heating -- pressing was carried out and it produced The metallic elements which can form above-mentioned carbon and an above-mentioned compound are specifically Si, Ti, V, Cr, Zr, Nb, Mo, Hf, Ta, and W. Si, Ti, Zr, and Hf are [among these] especially desirable. These metallic elements are 0.2-5. Weight % and twists 0.5-3 Weight % is desirable. Moreover, although the above-mentioned heating and pressing have the desirable inside of a vacuum, the reduced pressure or inert gas atmosphere which can suppress dissolution of the oxygen to the inside of a copper matrix is sufficient as them. Although it is desirable that it is beyond the copper melting point as for heating temperature, it should just be 800 degrees C or more. Since it depends for welding pressure on heating temperature, it is necessary to apply the welding pressure according to heating temperature. According to this method, dissolution into the copper of the above-mentioned metallic element can be suppressed, and a good volume phase can be formed in copper and a carbon fiber interface.

[0016] The copper-carbon fiber complex with the predetermined organization and predetermined property which were produced by such manufacture method is applicable to the part for which low-fever expansion and high temperature conduction are needed. Especially, since junction by silver solder with the board of copper-carbon fiber complex, nitriding aluminum, and an alumina is possible, it is applicable to the thermally conductive substrate of a semiconductor device.

[0017]

[Embodiments of the Invention]

(Example 1) The manufacture method of copper-carbon fiber complex is shown.

[0018] The pitch based carbon fiber was cut in length of 2-6mm, and Ti particle with a diameter of 10 micrometers or less was mixed to this. Furthermore, Cu particle of 100 or less meshes was mixed into the carbon fiber and Ti particle mixture. a mixed weight ratio -- a carbon fiber -- :Cu particle:Ti particle =9.54(g):36.4(g): -- it was 2.05 (g) Moreover, some were ground from the state [that a carbon fiber is cut by a series of mixtures]. They are 0.1mmt(s) about the mixture 1 of a carbon fiber, Cu particle, and Ti particle. It put in into the box (Cu container) 2 made with Cu board, and put into the heating pressing equipment shown in drawing 1 . However, drawing 1 shows the cross section of heating pressing equipment. The chamber 3 is formed in heating pressing equipment for atmosphere control, and the heating furnace 4 is installed in it. After surrounding mixture 1 with the side graphite jig 5, the up graphite board 6, the graphite board 7 with an up reservoir, the lower graphite board 8, and the graphite board 9 with a lower reservoir, it was put on the fluidity prevention pan 10, and was installed in the heating furnace 4. At this time, the lower pressurization fixture 11 for pressurization and the up pressurization fixture 12 were adjusted so that mixture 1 might be located in the center of a heating furnace 4. After installing and adjusting in the above-mentioned state, the inside of a chamber 3 was made into the degree of vacuum of 1×10^{-5} or more Torrs. In addition, the crevice between the Cu container 2 and the fixture made from a graphite was filled up with Cu grain.

[0019] Then, they are 20 kg/cm² with a pressurization fixture to mixture 1. Where a pressure is put, they are 1100 degrees C and 10min. Heating pressing was carried out. This produced the copper-carbon fiber complex on a disk with a diameter [of 65mm], and a thickness of 4mm.

[0020] Thus, as a result of observing with the scanning electron microscope (SEM) of the produced copper-carbon fiber complex, the volume phase of the gray to which the portions of a carbon fiber and white contrast exist [the portion of black contrast] in the interface of a copper matrix, and a carbon fiber and a copper matrix was observed. The elemental-analysis equipment (EDX) with which SEM is equipped analyzed each portion of a carbon fiber, a volume phase, and a copper matrix, and the result

shown in drawing 2, drawing 3, and drawing 4 was obtained. This shows that the volume phase which contains Ti, Cu, and carbon in the interface of a carbon fiber and a copper matrix is formed. Moreover, in the copper matrix, only the X-ray peak from copper is observed substantially. each element weight ratio of the produced copper-carbon fiber complex -- Cu:Ti:C=87.2:2.6:10.2 it was .

[0021] (Example 2) By the same manufacture method as the copper-carbon fiber complex shown in an example 1, copper-carbon fiber complex was produced so that it might become about 30 and 50 or 70% of carbon fiber volume fraction. It was made for the weight ratio of a carbon fiber and Ti particle to be set to about 4:1 at this time. Coefficient of thermal expansion was measured for the produced disc-like copper-carbon fiber complex between room temperature -300 degrees C. Moreover, thermal conductivity was measured with the laser flash method. The relation between coefficient of thermal expansion, and the thermal conductivity and carbon fiber volume fraction which were obtained is shown in drawing 5 and drawing 6, respectively. In order to attain both the coefficient of thermal expansion not more than $12 \times 10^{-6}/\text{degree K}$, and the thermal conductivity beyond 120 W/m degree K, 20 - 70% of a carbon fiber volume fraction is more suitable than these.

[0022] (Example 3) a carbon fiber volume fraction -- about 50% -- fixing -- the weight ratio of a carbon fiber and Ti particle -- about 4:1 and 2: -- it blended so that it might be set to 1 and 1:1, and copper-carbon fiber complex was produced using the same manufacture method as an example 1 The thermal conductivity of the produced copper-carbon fiber complex was measured with the laser flash method. Moreover, the average thickness of the volume phase containing Ti was measured from the SEM photograph. Then, the relation of the volume phase thickness and the thermal conductivity which are shown in drawing 7 was obtained. In order to obtain the thermal conductivity beyond 120 W/m degree K, the volume phase thickness of 2 micrometers or less is more suitable than this.

[0023] Moreover, when Ti and the weight ratio of a carbon fiber which were mixed to copper-carbon fiber complex, and the relation of thermal conductivity were arranged, Ti weight was set to Mm and the specific gravity of Mcf and Metal Ti was set to rho for a carbon fiber weight, the relation of drawing 8 was obtained. In order to obtain the thermal conductivity beyond 120 W/m degree K from this, the value of $Mm/(Mcf \times \rho)$ is 0.1. The following is suitable.

[0024] (Example 4) The value of about 50% and $Mm/(Mcf \times \rho)$ is a carbon fiber volume fraction About 0.05 Si, Zr, and Hf were blended, respectively so that it might change, and copper-carbon fiber complex was produced by the same method as an example 1. Such coefficient of thermal expansion and thermal conductivity became the value shown in Table 1. Si, Zr, and Hf can expect from this the effect which forms a volume phase in the interface of a carbon fiber and a copper matrix like Ti.

[0025]

[Table 1]

表 1

添加金属元素	熱膨張率 ($\times 10^{-6}/^{\circ}\text{K}$)	熱伝導度 ($\text{W}/\text{m}^{\circ}\text{K}$)
S i	7.2	175
Z r	8.4	212
H f	8.7	214

[0026] (Example 5) Silver solder was used on copper-carbon fiber complex with a diameter [of 65mm] produced in the example 1, and a thickness of 4mm, and the nitriding aluminum board of an one-side square (40mm and thickness 1mm) was joined. Junction is 3min(s) at 800 degrees C. It holds and is 3 degrees C/min. It cooled with the cooling rate. The crack was not observed by copper-carbon fiber complex and the zygote of nitriding aluminum by this junction method. Moreover, it can be said from there having been no curvature that it is in a good junction state. The -50-125-degree C thermal cycling test was carried out for the above-mentioned copper-carbon fiber complex and the carbonization aluminum zygote. Consequently, degradation of a zygote was not observed to 300 cycle.

[0027]

[Effect of the Invention] According to this invention, the copper-carbon fiber complex of low-thermal expansion and high temperature conduction is obtained by the low cost. Moreover, by using the copper-carbon fiber complex of this invention as a thermally conductive substrate of a semiconductor device, in the quality of a semiconductor device, it is reliable, and the semiconductor device excellent in thermolysis nature is obtained.

[Translation done.]

* NOTICES *

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2. **** shows the word which can not be translated.
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CLAIMS

[Claim(s)]

[Claim 1] Copper-carbon fiber complex characterized by the volume phase to which a copper matrix consists of copper substantially except for an unescapable impurity element, it has a metallic element in the interface of copper and a carbon fiber in the copper-carbon fiber complex which contains a carbon fiber in a copper matrix, and this metallic element reacted or dissolved to carbon, copper, or its both existing.

[Claim 2] Copper-carbon fiber complex characterized by for a copper matrix consisting of copper substantially except for an unescapable impurity element, having the metallic element which the melting point is higher than copper to the interface of copper and a carbon fiber, and forms carbon and a compound in it in the copper-carbon fiber complex which contains a carbon fiber in a copper matrix, and having the volume phase to which this metallic element reacted or dissolved to the aforementioned carbon, copper, or its both.

[Claim 3] In the copper-carbon fiber complex which contains a carbon fiber in a copper matrix A copper matrix consists of copper substantially except for an unescapable impurity element, and contains at least one metallic element in Si, Ti, V, Cr, Zr, Nb, Mo, Hf, Ta, and W in the interface of copper and a carbon fiber. Copper-carbon fiber complex characterized by having the volume phase to which this metallic element reacted or dissolved to the aforementioned carbon, copper, or its both.

[Claim 4] When the aforementioned metallic element sets the content of rho and carbon to Mcf for Mm and its specific gravity by the weight ratio by the weight ratio in either of the claims 1-3, the value of $Mm/(Mcf \times \rho)$ is 0.1. Copper-carbon fiber complex characterized by being the following.

[Claim 5] It is the copper-carbon fiber complex characterized by the aforementioned carbon fiber being 20 - 70% at the rate of volume in either of the claims 1-4.

[Claim 6] It sets to either of the claims 1-4, and the thickness of the aforementioned volume phase is 2.0 micrometers. Copper-carbon fiber complex characterized by being the following.

[Claim 7] Copper-carbon fiber complex with which coefficient of thermal expansion is characterized by less than $[12 \times 10^{-6} / \text{degree K}]$ and thermal conductivity being more than 120 W/m degrees K in either of the claims 1-6.

[Claim 8] In the manufacture method of the copper-carbon fiber complex which contains a carbon fiber in a copper matrix After mixing with a copper powder, a carbon fiber, and this carbon fiber the metallic element powder which forms a compound, The manufacture method of the copper-carbon fiber complex which carries out heating pressing and is characterized by making the volume phase to which the aforementioned copper matrix consisted of copper substantially except for the unescapable impurity element, and reacted or dissolved to carbon, copper, or its both to the interface of copper and a carbon fiber form.

[Claim 9] In the manufacture method of the copper-carbon fiber complex which contains a carbon fiber in a copper matrix After mixing the metallic element powder which the melting point is higher than a copper powder, a carbon fiber, and copper, and forms carbon and a compound, Carry out heating pressing and the aforementioned copper matrix consists of copper substantially except for an

unescapable impurity element. The manufacture method of the copper-carbon fiber complex characterized by making the volume phase to which the aforementioned metallic element powder with which the melting point is high with the powder and forms carbon and a compound from copper reacted or dissolved to the aforementioned carbon, copper, or its both form.

[Claim 10] In the manufacture method of the copper-carbon fiber complex which contains a carbon fiber in a copper matrix After mixing at least one metallic element powder in a copper powder, a carbon fiber and Si, Ti, V, Cr, Zr, Nb, Mo, Hf and Ta, and W powder, The manufacture method of the copper-carbon fiber complex which carries out heating pressing and is characterized by making the volume phase to which the aforementioned copper matrix changed from copper substantially except for the unescapable impurity element, and the aforementioned metallic element powder reacted or dissolved to the aforementioned carbon, copper, or its both form.

[Claim 11] Copper-carbon fiber complex with which coefficient of thermal expansion of this complex is characterized by less than $[12 \times 10^{-6} / \text{degree K}]$ and thermal conductivity being more than 120 W/m degrees K in the copper-carbon fiber complex which contains a carbon fiber in a copper matrix.

[Claim 12] Setting to the semiconductor device which carried the semiconductor device in the thermally conductive substrate, the aforementioned substrate is a semiconductor device which reaches and is characterized by the bird clapper from the copper-carbon fiber complex of a publication at either of 11 claim 1-7.

[Translation done.]